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(71)Applicant : NIPPON SYNTHETIC CHEM IND CO LTD:THE

(72)Inventor : NIMIYA KENJI
TOYOZUMI MASAHIKO
MASUMOTO HIROKI

(54) RESIN COMPOSITION, MOLDING PROCESS THEREOF AND MULTILAYER STRUCTURE THEREOF

(57)Abstract:

PROBLEM TO BE SOLVED: To provide a resin composition having a high melt moldability which prevents increase in resin pressure inside an extruder even at scrap return or continuous operation over a long time, a molding process thereof and a use thereof.

SOLUTION: This resin composition comprises a thermoplastic resin (A), an ethylene-vinyl acetate copolymer saponified product (B), an inorganic filler other than hydrotalcite compounds (C), a higher fatty acid metal salt (D) and, preferably, a hydrotalcite compound (E). Here, the average particle size of the inorganic filler (C) is 1 to 20 μm, the contents of (A), (B) and (C) are 30 to 99 wt.%, 0.5 to 20 wt.%, and 0.5 to 50 wt.%, respectively, against the total amount of (A), (B) and (C), and the content of (D) is 0.001 to 10 pts.wt. against 100 pts.wt. the total amount of (A), (B) and (C).

(E): 0.001 – 10 pts wt% based on total amount of (A)+(B)+(C)

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* NOTICES *

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Field of the Invention] the thermoplastics (A) with which, as for this invention, the long-run nature at the time of melting shaping has been improved, and an ethylene-vinylacetate copolymer saponification object (B) -- (-- it is hereafter written as EVOH.) -- it is further related with the shaping approach and multilayer-structure object using it about the resin constituent which consists of an inorganic filler (C) and a higher-fatty-acid metal salt (D).
[0002]
[Description of the Prior Art] although melting shaping of the mixture of the thermoplastics and EVOH which are represented by polyolefine system resin including polyethylene and polypropylene is carried out conventionally and various kinds of moldingses are obtained -- the object of this technique -- ** each -- it is divided roughly into two of the reuse (a scrap return or re grind) by recovery of acquire the physical properties which are not acquired , the kudzu of the product generated at the time of manufacture of the laminating structure of ** thermoplastics and EVOH and an edge , or a defective if independent . If it restricts to operation on industrial magnitude, the usefulness on industry has a remarkable direction in **.
[0003] However, the laminating structures which consist of the thermoplastics and EVOH like the above, such as a sheet and a film, are fabricated. The scrap constituent is used as a re grind layer. With melting shaping A film, When it is going to manufacture the laminating structures, such as a sheet, there is a trouble that cause gelation and the so-called long-run nature which the heat coloring resin with which this constituent is called burning at the time of shaping, and the carbonized resin adhere in an extruder, and cannot perform melting shaping continuously over a long period of time is inferior. Moreover, in order that the phase separation foreign matter of the gelation object, the thermoplastics, and EVOH at the time of this melting may often mix into a moldings, in film shaping, it becomes the big cause of the defect of moldingses including generating of a fish eye, and debasement of a product is not escaped.
[0004] In recent years, it aims at improvement in the rigidity of the laminating structure, thermal resistance, and an appearance especially. Although blending an inorganic filler with thermoplastics is performed and melting shaping of the resin constituent which consists of the thermoplastics (A), EVOH (B), and the inorganic filler (C) which carried out recovery reuse of the laminating structure will be performed In this melting fabrication operation, in a short time, the resin pressure force inside an extruder goes up, and operating becomes impossible. The phenomenon it becomes impossible so-called to long-run fabricate is accepted notably, and it is forced the very troublesome activity that an extruder must be disassembled for whenever [the / every] and exchange or the screw of a screen pack must be cleaned.
[0005] Then, as the above-mentioned solution, by JP,3-72541,A, it consists of EVOH beyond 20 mol % whenever [inorganic substance / which are chosen from EVOH, the titanium oxide, the talc, the calcium carbonate, mica, and absorptivity inorganic substance beyond 96 mol % whenever / polyolefine and

ethylene content % and saponification / at least one sort of /, and ethylene content % and saponification], and excels in compatibility, and the resin constituent effective in re grind is proposed. [of 20-65 mols] [of 68-98 mols]
[0006]
[Problem(s) to be Solved by the Invention] However, the result to which this invention person etc. examined the above-mentioned official report disclosure technique in the detail, Although the effectiveness of the appearance of a moldings and reduction of a phase separation foreign matter which are obtained with this technique is accepted to some extent About control of the resin pressure force at the time of a melting fabricating operation, detailed examination was not carried out, but the phenomenon the resin pressure force inside the extruder at the time of carrying out melting shaping goes up, and it becomes impossible operating was seen, and it became clear that it is still inadequate.
[0007] So, in this invention, while excelling in an appearance or thermal resistance under such a background, also in continuous running of a repeated scrap return and repeated long duration, the resin pressure force inside an extruder does not go up, but it aims at offering the resin constituent excellent in the melting moldability.
[0008]
[Means for Solving the Problem] Then, the result to which this invention person etc. repeated research wholeheartedly in view of the above-mentioned situation, It consists of the inorganic fillers (C) and higher-fatty-acid metal salts (D) other than a (Thermoplastics A) EVOH(B) hydrotalcite system compound. And the mean particle diameter of an inorganic filler (C) is 1-20 micrometers, and the total quantity of thermoplastics (A), EVOH (B), and an inorganic filler (C) is received. The content of an inorganic filler (C) 0.5 to 20% of the weight 30 to 99% of the weight at 0.5 - 50 % of the weight [the content of thermoplastics (A)] [the content of EVOH (B)] Furthermore, the resin constituent whose content of a higher-fatty-acid metal salt (D) is 0.001 - 10 weight section completed a header and this invention for agreeing with the above-mentioned object to the total quantity 100 weight section of thermoplastics (A), EVOH (B), and an inorganic filler (C).
[0009] In this invention, further, when it is the resin constituent which comes to carry out 0.001-10 weight section content of the hydrotalcite system compound (E) to the total quantity 100 weight section of thermoplastics (A), EVOH (B), and an inorganic filler (C), and when it is the resin constituent which satisfies following ** type, the effectiveness of this invention is demonstrated notably.
[Equation 2] WbxDcxWc<=4000 ... Content of the ethylene-vinylacetate copolymer saponification object (B) to the total quantity of (A) in a **Wb:resin constituent, (B), and (C) (% of the weight)
Dc: Mean particle diameter of an inorganic filler (C) (micrometer)
Wc: The content of the inorganic filler (C) to the total quantity of (A) in a resin constituent, (B), and (C) (% of the weight)
[0010] Moreover, the resin constituent of this invention is useful also on the multilayer-structure object which has the configuration of the resin constituent layer / adhesion resin layer / EVOH layer of a thermoplastics layer / this invention, the resin constituent layer / adhesion resin layer / EVOH layer / adhesion resin layer / thermoplastics layer of a thermoplastics layer / this invention, the resin constituent layer of a thermoplastics layer / this invention / a resin constituent layer / thermoplastics layer of an adhesion resin layer / EVOH layer / adhesion resin layer / this invention.
[0011]
[Embodiment of the Invention] Below, this invention is explained at a detail. Although especially the thermoplastics (A) of this invention is not restricted but polyolefine system resin, polyamide system resin, polyester system resin, polyether system resin, polyurethane system resin, polycarbonate system resin, etc. are mentioned, polyolefine system resin is used preferably especially. Moreover, these may be used independently, or they may be used for two sorts, using together. Although especially polyolefine system resin is not limited, it becomes able [the chlorine which is manufactured using a Ziegler type catalyst and originates in a catalyst] to acquire the effectiveness of this invention more notably by using the polyolefine system resin contained 3-150 ppm preferably 1-300 ppm.
[0012] As this polyolefine system resin, the various polyethylene of high density, semi-gross density,

and a low consistency, Homopolymers, such as polypropylene, polybutene, and the poly pentene, ethylene propylene rubber, Ethylene or a propylene is made into a subject. A copolymer with a with a carbon numbers [, such as 1-butene and 1-hexene,] of about two to 20 alpha olefin, Furthermore, the olefin-vinyl acetate copolymer which has the presentation comparatively near polyolefine whose content of olefins, such as ethylene or a propylene, is more than 90 mol %, Independent or a thing which carried out graft denaturation of the copolymer with unsaturated carboxylic acid etc. of the above-mentioned or polyolefine system resin, such as ionomer resin with which the olefin-(meta) acrylic ester copolymer and the metal ion were embellished, etc. is usable to one sort or two or more sort arbitration. Especially among these, in the activity of the polyethylene system resin for 0.1 - 15g / 10 minutes (190 degrees C, 2160g), or the polypropylene regin for 0.1 - 12g / 10 minutes (230 degrees C, 2160g), it is easy to generate the problem of resin pressure force lifting, and the melt index (MI) is excellent also in the effectiveness of this invention.

[0013] as EVOH (B) used for this invention -- ethylene content 10 - 70-mol % -- more than 90 mol % and the thing which has the presentation beyond 95 mol % preferably are mentioned whenever [saponification / of a 20 - 60 mol % and vinyl acetate part] preferably. Less than [ethylene content 10 mol %], since thermal stability is bad, a melting moldability falls, and oxygen cutoff nature falls when an ethylene content exceeds 70-mol %, it becomes lacking in practicability. Moreover, since thermal stability is poor and less than [90 mol %] inferior to physical properties, such as oxygen cutoff nature, oilproof, and a water resisting property, whenever [saponification / of a vinyl acetate part] is lacking in practicability.

[0014] Above EVOH (B) is ethylene and vinyl acetate. (or vinyl alcohol which saponified it) In others, the 3rd component, such as vinyl ester other than alpha olefins, such as unsaturated carboxylic acid, its ester or a salt, a partial saturation sulfonic acid or its salt, acrylamide (meta), acrylonitrile (meta), a propylene, a butene, alpha-octene, and alpha-octadecene, and vinyl acetate, may be by little **** not more than about 10 mol %.

[0015] As an inorganic filler (C), especially if it is things other than a hydrotalcite system compound, it will not be limited, for example, a mica, talc, a calcium carbonate, titanium oxide, a kaolin, clay, a glass flake, glass BISU, a vermiculite, a smectite, etc. are mentioned, it may be used independently or two or more sorts may be used together. In this invention, it is required for the mean particle diameter of this inorganic filler (C) to be 1-20 micrometers, and it is 5-15 micrometers especially preferably 3-18 micrometers preferably. In less than 1 micrometer, if the gel according [this mean particle diameter] to condensation of this particle will occur in a moldings, and the heat-resistant rigidity of the laminating structure will also run short of it and it exceeds 20 micrometers, it will become difficult to control lifting of the resin pressure force. In addition, the mean particle diameter said here is a value measured by the well-known measuring method, for example, a light transmission centrifuge etc.

[0016] Especially as a higher-fatty-acid metal salt (D), although not limited, a with a carbon numbers of eight or more higher-fatty-acid metal salt is desirable. For example, a lauric acid, a tridecyl acid, a myristic acid, pentadecyl acid, A palmitic acid, a heptadecyl acid, stearin acid, hydroxy stearin acid, Alkaline-earth-metal salts, such as alkali-metal salts, such as sodium salt of higher fatty acids, such as a nonadecane acid, oleic acid, a capric acid, behenic acid, and linolic acid, and potassium salt, magnesium salt, a calcium salt, and barium salt, a zinc metal salt, etc. are mentioned. The metal salt of stearin acid, oleic acid, and a lauric acid is remarkable especially in respect of effectiveness also in this inside.

[0017] Although it consists of the above-mentioned thermoplastics (A), EVOH (B), an inorganic filler (C), and a higher-fatty-acid metal salt (D) in this invention About the content of each of this component, the total quantity of thermoplastics (A), EVOH (B), and an inorganic filler (C) is received. The content of thermoplastics (A) preferably 30 to 99% of the weight 40 - 98 % of the weight, It is 50 - 93 % of the weight preferably. The content of EVOH (B) Furthermore, 0.5 - 20 % of the weight, It is 2 - 12 % of the weight still more preferably, and the content of an inorganic filler (C) is 5 - 30 % of the weight still more preferably one to 40% of the weight preferably 0.5 to 50% of the weight one to 16% of the weight.

[0018] The content of this thermoplastics (A) becomes difficult [it / to control lifting of the resin pressure force at less than 30 % of the weight], the appearance of mold goods also gets worse further,

and when 99 % of the weight is exceeded, the gas barrier nature of the laminating structure and heat-resistant rigidity will run short. If the gas barrier nature of the laminating structure will run short of the contents of EVOH (B) at less than 0.5 % of the weight and 20 % of the weight is exceeded, it will become difficult to control lifting of the resin pressure force, and the appearance of mold goods will also get worse further. If the heat-resistant rigidity of the laminating structure will run short of the contents of an inorganic filler (C) at less than 0.5 % of the weight and 50 % of the weight is exceeded, it will become difficult to control lifting of the resin pressure force, and the appearance of mold goods will also get worse further.

[0019] furthermore, the content of a higher-fatty-acid metal salt (D) -- the total quantity 100 weight section of thermoplastics (A), EVOH (B), and an inorganic filler (C) -- receiving -- 0.001 - 10 weight section -- desirable -- 0.005 - 5 weight section -- it is 0.01 - 1 weight section still more preferably. When the content of this higher-fatty-acid metal salt (D) becomes difficult [it / to control lifting of the resin pressure force under in the 0.001 weight section] and 10 weight sections are exceeded, the appearance of a moldings will get worse and commodity value will fall.

[0020] Furthermore, in this invention, in addition to the component of above-mentioned (A) - (D), it is desirable to make a hydrotalcite system compound (E) contain, and the melting moldability of this invention improves much more by content of this hydrotalcite system compound (E). As a hydrotalcite system compound (E) For example, they are a general formula and the compound shown by MxAly (OH)2x+3y-2z(E) z-aH2O (a positive number and a are [the inside M of a formula / Mg, calcium, or Zn and E] 0 or a positive number for CO3 or HPO 4 and xy, and z). Specifically Mg4.5aluminum2 (OH) 13CO3.3.5H2O, Mg5aluminum2 (OH) 14CO3and4H2O, Mg6aluminum2 (OH) 16CO3and4H2O, Mg8aluminum2 (-- OH --) -- 20 -- CO -- 3.5 -- H -- two -- O -- Mg -- ten -- aluminum -- two -- (-- OH --) -- 22 (CO3) -- 2.4 -- H -- two -- O -- Mg -- six -- aluminum -- two -- (-- OH --) -- 16 -- HPO -- 4.4 -- H -- two -- O -- calcium -- six -- aluminum -- two -- (-- OH --) -- 16 -- CO -- three - four -- H -- two -- O -- Zn -- six -- aluminum -- six -- (-- OH --) -- 16 -- CO -- three - four -- H -- two -- O -- etc. etc. -- mentioning -- having . Moreover, equivalent effectiveness is expectable even if a part of OH for example, not only the above but in Mg2aluminum(OH)9.3H2O is what it is not clearly indicated as and the thing (a= 0) from which water of crystallization was removed further of the **** chemical formula permuted by CO3 or HPO4. Effectiveness with the most remarkable compound whose E M is CO3 in Mg among these is shown especially.

[0021] Furthermore, it is also possible to use the hydrotalcite system solid solution shown by the following general formula as a hydrotalcite system compound (E).

[[(M12+) y1(M22+) y2]1-xMx3+(OH) 2 An-x/n-mH2O]

(The metal chosen from Mg, calcium, Sr, and Ba at least one sort of M12in formula+) For An-, Mx3+ is [the anion of n **, x, and y1, y2 and m] the metal with which M22+ is chosen from Zn, Cd, Pb, and Sn, and trivalent metal and the positive number shown by 0< x<=0.5, 0.5<y1<1, y1+y2=1, and 0<=m<2, respectively.

In the above-mentioned general formula, as M12+, Mg and calcium are desirable, Zn and Cd are desirable as M22+, and although aluminum, Bi, In, Sb, B, Ga, Ti, etc. can be further illustrated as Mx3+, aluminum is practical. moreover -- An - ****** -- CO -- 32 - OH - HCO -- three - a salicylic acid -- ion -- a citric acid -- ion -- a tartaric acid -- ion -- NO -- three - I - two (OOC-COO) - ClO -- four - CH -- three -- COO - CO -- 32 - two (OOCHC=CHCOO) - [-- Fe -- (-- CN --) -- six --] -- four - mentioning -- having -- CO32- and OH- being useful .

[0022] As a concrete example of this hydrotalcite system solid solution [Mg0.75 Zn0.25] 0.67 aluminum0.33 (OH) 2(CO3) 0.165.0.45H2O, [Mg0.79 Zn0.21]0.7 aluminum0.3(OH)2(CO3)0.15, Mg1/7 calcium3 / [7 Zn3/7] 0.7 aluminum0.3(OH) 2 (OOCHC=CHCOO)0.15.0.41H2O, Mg6 / [7 Cd1/7]0.7 aluminum0.3 (OH) 2(CH3COO) 0.3.0.34H2O, Mg5 / [7 Pd2/7] 0.7 aluminum0.30 (OH) 2 (CO3) 0.15.0.52H2O, [Mg0.74 Zn0.26]0.68 aluminum0.32(OH)2(CO3)0.16, [Mg0.56 Zn0.44] 0.68 aluminum0.32(OH) 2 (CO3)0.16.0.2H2O, [Mg0.81 Zn0.19]0.74 aluminum0.26(OH)2(CO3)0.13, [Mg0.75 Zn0.25] 0.8 aluminum0.20(OH) 2 (CO3)0.10.0.16H2O, [Mg0.71 Zn0.29]0.7 aluminum0.30 (OH)2(NO3)0.30, [Mg0.71 Zn0.29]0.7 aluminum0.30(OH)2(OOCHC=CHCOO)0.15, and [Mg0.14

calcium0.57 Zn0.28]0.7 aluminum0.30 (OH) 2.3.0.25H2O etc. is mentioned. [Mg0.75 Zn0.25] 0.67 aluminum0.33 (OH) 2(CO3) 0.165.0.45H2O, [Mg0.79 Zn0.21]0.7 aluminum0.3(OH)2(CO3)0.15, Mg6 / [7 Cd1/7] 0.7 aluminum0.3(OH) 2 (CH3COO)0.3.0.34H2O, Mg5 / [7 Pd2/7] 0.7 aluminum0.30(OH) 2 (CO3)0.15.0.52H2O is used suitably.

[0023] As for the content of this hydrotalcite system compound (E), it is desirable that it is 0.001 - 10 weight section to the total quantity 100 weight section of thermoplastics (A), EVOH (B), and an inorganic filler (C), and it is especially desirable further 0.005 - 5 weight sections, and that it is 0.01 - 1 weight section. If it may become inadequate [under the 0.001 weight section] controlling [of resin pressure force lifting] the content of this hydrotalcite system compound (E) and 10 weight sections are exceeded, the appearance of a moldings will get worse, commodity value will fall, and it is not desirable.

[0024] Moreover, although this invention is a resin constituent which consists of (A) - (D) or (A) - (E) like the above, when satisfying above-mentioned ** type especially, it shows the especially excellent re grind nature and a melting moldability. Controlling [of resin pressure force lifting] may become inadequate, and it stops demonstrating the effectiveness of this invention notably then in a ** ** type, so that the value of WbxDcxWc may exceed 4000. Especially in order to satisfy a ** ** type, it is desirable to lessen the content of EVOH (B) and/or the content of an inorganic filler (C).

[0025] Although the resin constituent of this invention comes to blend (A) - (D) or (A) - (E) like the above, any are sufficient as carrying out package combination of (A) - (E), and carrying out sequential combination of the component which remains, after not being limited but blending two components of arbitration etc., and especially the combination approach is chosen suitably and adopted. It is desirable to blend beforehand a higher-fatty-acid metal salt (D) and/or a hydrotalcite system compound (E) with thermoplastics (A) even especially in inside at the point which can control generating of a foreign matter like the gel of a moldings.

[0026] The mode of arbitration is mentioned as this mixed means. For example, [whether thermoplastics (A), a higher-fatty-acid metal salt (D) or thermoplastics (A) and a higher-fatty-acid metal salt (D), and a hydrotalcite system compound (E) are mixed with a Henschel mixer, a tumbler, etc., and] How to carry out melting mixing of the inorganic filler (C), and carry out melting mixing of the EVOH (B) further, after carrying out melting mixing with an extruder etc., Furthermore, the approach of carrying out melting mixing of the laminating structure more than two-layer [of the layer which consists of mixture of the layer which consists of EVOH (B), thermoplastics (A), an inorganic filler (C), and a higher-fatty-acid metal salt (D) (further hydrotalcite system compound (E))] again etc. is mentioned. In the latter approach, the approach of carrying out melting mixing of the crushing articles (the so-called re grind), such as kudzu usually generated at the time of manufacture of the above-mentioned laminating structure, an edge, and a defective, etc. is mentioned.

[0027] The resin constituent of this invention obtained in this way is used for various kinds of melting moldingses. In manufacture of this melting moldings, it is desirable that it is desirable to consider as about 160-280 degrees C as temperature conditions at the time of melting shaping, and the screen pack of at least one or more sheets is used in carrying out melting shaping with an extruder, and there is opening of the screen by 50 micrometers or more, and it is desirable that it is further 100-400 micrometers. It becomes [controlling / of resin pressure force lifting / this opening] insufficient [less than 50 micrometers] and is not desirable.

[0028] On the occasion of shaping, suitable combination of the well-known additives, such as fillers other than lubricant, such as reinforcing materials, such as a glass fiber and a carbon fiber, low molecular weight polyethylene, low molecular weight polypropylene, paraffin, a higher-fatty-acid AMAIDO system, and an epoxy system, and the above-mentioned inorganic filler (C), a coloring agent, an antioxidant, an ultraviolet ray absorbent, an antimicrobial agent, and a foaming agent, may be carried out if needed.

[0029] As a melting fabricating method, methods of fabricating arbitration, such as the injection-molding method, compression forming, and an extrusion method, are employable. among these -- as an extrusion method -- the T-die method, a blow molding method, a pipe extrusion method, and a line -- an

extrusion method, a variant die extrusion method, a tubular film process, the melt span method, etc. are mentioned.

[0030] The configuration of the moldings obtained by the approach of this invention is arbitrary. A film, It is also important to consider as the multilayer-structure object to carry out. a sheet, a tape, a bottle, a tube, a tank, a hose, a pipe, a filament, a variant cross-section extrusion object, etc. -- accepting it -- not becoming -- the resin constituent of this invention -- at least -- much more -- ** -- As other party resin in the case of carrying out a laminating, polyolefine system resin, EVOH, nylon 6, nylon 6, the polyamide system resin of 6 grades, vinylidene-chloride system resin, styrene resin, polyester system resin, etc. are often used. Of course, even if it is the usual thermoplastics other than the above, for example, a polycarbonate, vinyl chloride system resin, acrylic resin, vinylester resin, a polyester elastomer, a polyurethane elastomer, chlorinated polyethylene, and chlorination polypropylene, it does not interfere at all. The above-mentioned thermoplastics (A) and the same thing as EVOH (B) can be used for this thermoplastics and EVOH.

[0031] As lamination of a concrete multilayer-structure object, the resin constituent layer / adhesion resin layer / EVOH layer of a thermoplastics layer / this invention, The resin constituent layer / adhesion resin layer / EVOH layer / adhesion resin layer / thermoplastics layer of a thermoplastics layer / this invention, the resin constituent layer of a thermoplastics layer / this invention / the resin constituent layer / thermoplastics layer of an adhesion resin layer / EVOH layer / adhesion resin layer / this invention -- further -- the resin constituent layer / adhesion resin layer / EVOH layer of this invention -- The resin constituent layer / adhesion resin layer / EVOH layer / adhesion resin layer / EVOH layer of this invention, The resin constituent layer / adhesion resin layer / EVOH layer / adhesion resin layer / thermoplastics layer of this invention, the resin constituent layer of this invention / a resin constituent layer / thermoplastics layer of an adhesion resin layer / EVOH layer / adhesion resin layer / this invention is mentioned.

[0032] As adhesion resin used for this adhesion resin layer The ethylene-alpha olefin copolymer of the consistency 0.86 which could use well-known adhesives, for example, denaturalized with unsaturated carboxylic acid or its anhydride - 0.95 g/cm3 is desirable. The above-mentioned polyolefine system resin and the same resin can be obtained copolymerization or by carrying out graft denaturation with unsaturated carboxylic acid or its anhydride, and, of course, the blend of a non-denaturalized ethylene-alpha olefin copolymer, unsaturated carboxylic acid, or its anhydride is also included in denaturation. As unsaturated carboxylic acid or its anhydride, a maleic acid, a maleic anhydride, a fumaric acid, an acrylic acid, a methacrylic acid, a crotonic acid, an itaconic acid, a citraconic acid, hexahydro phthalic anhydride, etc. are mentioned, and a maleic anhydride is used suitably especially.

[0033] 0.001 - 10 % of the weight is desirable still more desirable, and the unsaturated carboxylic acid contained in the ethylene-alpha olefin copolymer at this time or its amount of anhydrides is 0.01 - 5 % of the weight. It worsens [a lifting and a moldability] crosslinking reaction and is not desirable if many [if there are few these contents, adhesive strength will decline, and / conversely]. It is also possible to mix this adhesive resin with the adjoining layer.

[0034] Not only the shape of a sheet or a film but the thing which it can fabricate in containers, such as the shape of a pipe tube and a tank bottle, etc., and this multilayer-structure object is again heated further at about 100-150 degrees C by a coextrusion process, a coinjection-molding method, the above-mentioned co-extrusion inflation fabricating method, an above-mentioned blow molding method, etc., and is extended by the blow extending method etc. is possible for the multilayer-structure object of this invention. Moreover, on each class (except the resin constituent layer of this invention) of the multilayer-structure object of this invention, an antioxidant, lubricant, an antistatic agent, a plasticizer, a coloring agent, an ultraviolet ray absorbent, an antimicrobial agent, inorganic, an organic filler, etc. can also be added in the range which does not check the effectiveness of this invention for [, such as fabricating-operation nature and physical properties,] improvement.

[0035]

[Example] An example is given to below and this invention is concretely explained to it. In addition, especially, it means weight criteria that it is with the "section" and "%" among an example, as long as

there is no notice. The following resin and compounds were prepared.
[Polyolefine system resin (A)]
A1; polypropylene (MI=0.8g /, 10 minutes, consistency 3, the chlorine content of 110 ppm of 0.90g/cm)
A2; maleic-anhydride denaturation polypropylene (MI=1.0g /, 10 minutes, consistency 3, the chlorine content of 80 ppm of 0.89g/cm)
A3; high density polyethylene (MI=6.0g /, 10 minutes, consistency 0.952 g/cm3, chlorine content of 15 ppm)
In addition, above-mentioned MI expresses the melt flow index at the time of 230 degrees C (polypropylene), 190 degrees C (polyethylene), and 2160g load.
[0036] [EVOH(B)]
B1; 98.7-mol %, MI=8g / still more nearly above-mentioned MI expresses [whenever / ethylene content % and saponification / 99.6 mol % and MI =3.5g /, and whenever / 10 minute B-2; content / ethylene / % and saponification] the melt flow index at the time of 210 degrees C and 2160g load for 10 minutes 99.5-mol % and MI =6g /, and whenever [10 minute B3; content / ethylene / % and saponification]. [of 38 mols] [of 27 mols] [of 40 mols]
[0037] [An inorganic filler (C)]
C1; talc (mean particle diameter of 11 micrometers, talc powder PK-C (made in [shrine] formation [Wood]))
C2; kaolin (mean particle diameter of 0.4 micrometers, ASP200 (made in [shrine] formation [Wood]))

C3; talc (mean particle diameter of 22 micrometers, crown talc DR (Matsumura industrial company make))
[Higher-fatty-acid metal salt (D)]
D1; calcium stearate (Nippon Oil & Fats Co., Ltd. make)
D2;12-hydroxy magnesium stearate (made in [shrine] formation [Yoshinobu])
[0038] [Hydrotalcite system compound (E)]
E1;Mg4.5aluminum2(OH)13CO3.3.5H2OE2; [Mg0.75 Zn0.25]0.67 aluminum0.33 (OH) 2(CO3) 0.165.0.45H2O [0039] [Adhesion resin]
F1; denaturation polyolefine system resin (trade name: Modic AP P512 (Mitsubishi Chemical make))
[0040] Melting mixing of the **** thermoplastics (A) shown in one to examples 1-3 and example of comparison 7 table 1, EVOH (B), an inorganic filler (C), a higher-fatty-acid metal salt (D), and the hydrotalcite system compound (E) was carried out with the single screw extruder, melting shaping was performed, and the sheet was obtained. The process condition is as follows.
Process condition ** It comes out. Machine : The diameter single screw extruder of 40mm A screw : ratio-of-length-to-diameter=28 Compression ratio 3.5 Screen pack : It is used two opening 120micrometer things. DA I : A coat hanger type ** It comes out. ** Whenever : 240 degrees C of cylinder points die 230degree C screw-speed: -- actuation of having ground 40rpm profit **** sheet and performing sheet forming on these conditions again was repeated 10 times, and change of the resin pressure at the time of the 1st time, the 5th time, and the 10th shaping was measured.
[0041] Moreover, the appearance of the 1st time, the 5th time, and the 10th obtained sheet was observed visually, and the following valuation bases estimated.
** Surface smooth nature O ... The surface deterioration of a stripe or a front face is hardly accepted.
** ... The surface deterioration of a stripe or a front face is accepted a little.
x ... Many surface deterioration of a stripe or a front face is accepted.
** The number of the gel foreign matter of 0.4mm or more of diameters of per gel foreign matter sheet 100cm2 (10cmx10cm) was measured, and the following criteria estimated.
O ... Less than [two piece] ** ... 3 - 10 piece x ... 11 or more of each result are shown in a table 2.
[0042]
[A table 1]
(A) (B) (C) (D) (E) ** type (Loadings) (loadings) (loadings) (particle size) (loadings) (loadings) Value
Example 1 A1 A2 B-2 C1 11 micrometers D2 E1 528 (78) (6) (4) (12) (0.2) (0.2) ** 2 A3 B3 C1 11

micrometers D1 E2704 (84) (8) and (8) (0.3) (0.1) ** 3 A1 A2 B1 C1 11 micrometers D1 --- 1760 (64) (10) (10) (16) (0.5) Example 1 of a comparison A1 B1 C1 11 micrometers D1 --- 12672 (28) (24) (48) (0.5) ** 2 A1 A2 B1 C1 11micrometer D1 --- 5280 (44) (10) (30) (16) (0.5) ** 3 A1 A2 B1 C1 11 micrometers D1 --- 6600 (20) (10) (10) (60) (0.5) ** 4 A1 A2 B1 C1 11 micrometers --- --- 1760 (64) (10) (10) (16) ** 5 A1 A2 B1C1 11 micrometers D1 --- 1760 (64) (10) (10) (16) (15) ** 6 A1 A2 B1 C222micrometer D1 --- 3520 (64) (10) (10) (16) (0.5) ** 7 A1 A2 B1 C3 0.4 micrometers D1--- 64 (64) (10) (10) (16) (0.5) The loadings of notes (A), (B), and (C) receive the total quantity of (A), (B), and (C). It is ******% and the loadings of (D) are total quantity 100 weight of (A), (B), and (C). It is the weight section to the section.

[0043]

[A table 2]

The 1st time The 5th time The 10th time Resin pressure force Appearance Resin pressure force Appearance Resin pressure force Appearance ** ** (kg/cm2) (kg/cm2) ** ** (kg/cm2) ** ** example 1 70 O O 71 O O 72 O O ** 2 68 O O 68 O O 69 O O ** 3 75 OO 76 O Example 1 of an O 76 ** ** comparison 81 x ** (*) (*) and (*) (*) (*) (*)

** 2 75 ** ** 98 x x (*) (*) (*)

** 3 80 x O (*) (*) and (*) (*) (*) (*)

** 4 76 O O 89 ** ** 102 x x ** 5 70 x O 70 x O 70 x ** ** 6 77 ** O 87 x ** 100 x x ** 7 75 O x 77 O x 78 ** x (*) : [Since the resin pressure force is too high] It could not measure and a sheet was not obtained, either.

[0044] As opposed to the mixture 100 weight section of 80% (A1) of example 4 polyolefine system resin, and 20% of inorganic fillers (C1) Resin constituent (a) obtained by carrying out melting mixing of the higher-fatty-acid metal salt (D1) 0.5 section and the hydrotalcite system compound (E2) 0.5 section with a single screw extruder is used. resin constituent (**) -- a layer / resin constituent (**) -- a layer / adhesion resin (F1) -- a layer / EVOH (B1) layer / adhesion resin (F1) layer / resin constituent (**) -- the configuration of a layer (thickness = 100 micrometers / 400 micrometers / 100 micrometers / 100 micrometers / 100 micrometers / 500 micrometers) The multilayer-structure object which it has was manufactured on the following conditions using the four-sort six-layer feed block die.

[0045]

Process condition ** It comes out. Diameter extruder of machine **65mm (for polyolefine system resin (A1) layers) ** The diameter extruder of 65mm (resin constituent (**) or (**) for layers) ** The diameter extruder of 30mm (for adhesive resin (F1) layers) ** Diameter extruder of 30mm (for EVOH (B1) layers) Extrusion temperature C1 C2 C3 C4 AD FB Die (degree C) ** 200 230 230 210 220 220 230 ** 200 230 230 210 220 220 230 ** 200 230 --- --- 220 220 230 ** 200 230 --- --- 220 220 230 Screw speed **20rpm, **25rpm, **10rpm, **15rpm Die width of face 650mm Extrusion outlet 24kg/hr Taking over rate 0.33 m/min Roll temperature 80 degrees C [0046] Again resin constituent (b) which pulverized the multilayer-structure object acquired above to 1-5mm angle extent on these conditions as a layer of the above-mentioned ** A multilayer-structure object, resin constituent (**) -- a layer / resin constituent (**) -- a layer / adhesion resin (F1) -- a layer / EVOH (B1) layer / adhesion resin (F1) layer / resin constituent (**) -- the layer (thickness = 100 micrometers / 400 micrometers / 100 micrometers / 100 micrometers / 100 micrometers / 500 micrometers) was manufactured. The cup was fabricated with a vaccum pressure sky making machine on the following conditions using the multilayer-structure object which repeated this 10 times as the 1st time (scrap return), carried out this actuation, and was acquired by the 1st time, the 5th time, and the 10th time. In addition, the EVOH content in the 1st resin constituent (b) layer of a multilayer-structure object was about 9%, and the 5th time and the 10th EVOH content were about 13%. Moreover, the content of an inorganic filler was [about 0.4 sections, 5th about 0.3 sections / 10th /, and hydrotalcite system compound of higher-fatty-acid metal salt] the 1st about 0.4 sections and the 5th about 0.3 sections [10th] the time [10th] the 5th time and about 14% the time [1st] about 16%.

[0047] process condition heater temperature 450 degrees-C layered product skin temperature of vertical 160-degree-C cup configuration regio-oralis; -- 9x9cm and pars-basilaris-ossis-occipitalis; -- the

following way estimated the appearance of the cup obtained depth;6.5cm 8x8cm.
[0048] (Appearance)
Visual observation of the appearance of the obtained cup was carried out, and the following criteria estimated.
--- surface smooth nature -- good -- a stripe and foreign-matter-less b --- some -- foreign-matter[those with a stripe, and]-less c --- overall -- a stripe -- many foreign-matter-less d --- overall -- a stripe -- many -- some -- those [e] with a foreign matter --- overall -- a stripe and a foreign matter -- many
[0049] In example 5 example 4, except having used the resin constituent obtained to the mixture 100 section of 85% (A1) of polyolefine system resin, and 15% of inorganic fillers (C1) by carrying out melting mixing of the higher-fatty-acid metal salt (D1) 0.8 section and the hydrotalcite system compound (E1) 0.3 section with a single screw extruder as resin constituent (**), it carried out similarly and evaluated similarly.
[0050] example 6 example 4 -- setting -- the configuration of a multilayer-structure object -- resin constituent (**) -- a layer / resin constituent (**) -- except having been referred to as a layer / adhesion resin (F1) layer / EVOH (B1) layer / adhesion resin (F1) layer / resin constituent (b) layer / resin constituent (a) layer =200micrometer / 200 micrometers / 100 micrometers / 100 micrometers / 100 micrometers / 200 micrometers / 400 micrometers, it carried out similarly and evaluated similarly.
[0051] In example of comparison 8 example 4, except having used only the polyolefine system resin (A1) of an example 4, and the mixture of an inorganic filler (C1) as resin constituent (**), it carried out similarly and evaluated similarly.
[0052] In example of comparison 9 example 4, except having used only the polyolefine system resin (A1) of an example 5, and the mixture of an inorganic filler (C1) as resin constituent (**), it carried out similarly and evaluated similarly. Each result is shown in a table 3.
[0053]
[A table 3]

	外観		
	1回目	5回目	10回目
実施例4	a	b	b
〃 5	a	a	a
〃 6	a	b	b
比較例8	b	d	e
〃 9	a	d	e

注）上記回数は、スクラップリターンの回数を表す。

[0054]
[Effect of the Invention] (A) - (D) component like the above, since it blends (A) - (E) component preferably and (C) has specific mean particle diameter further, while the resin constituent of this invention is excellent in an appearance or thermal resistance The resin pressure force inside an extruder does not go up in continuous running of a repeated scrap return or repeated long duration. It is the resin constituent excellent in the melting moldability, and it can use as various multilayer-structure objects, and is useful on a package film, a container, a bottle bottle, a food tray, a sheet, various device components, etc.

[Translation done.]